Enhancement of magnetic ordering by the stress fields of grain boundaries in ferromagnets.

A. Kadigrobov*[1,2], Z. Ivanov[1]
[1] Department of Microelectronics and Nanoscience, Chalmers University of Technology and Göteborg University, S-412 96 Göteborg, Sweden
[2] B. I. Verkin Institute for Low Temperature Physics & Engineering, National Academy of Science of Ukraine, 47 Lenin Ave., 310164 Kharkov, Ukraine

R. I. Shekhter[3], and M. Jonson[3]
[1] Department of Applied Physics, Chalmers University of Technology and Göteborg University, SE-412 96 Göteborg, Sweden
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In the paper we predict a distinctive change of magnetic properties and considerable increase of the Curie temperature caused by the strain fields of grain boundaries in ferromagnetic films. It is shown that a sheet of spontaneous magnetization may arise along a grain boundary at temperatures greater than the bulk Curie temperature. The temperature dependence and space distribution of magnetization in a ferromagnetic film with grain boundaries are calculated. We found that 45° grain boundaries can produce long-range strain fields that results in the width of the magnetic sheet along the boundary of the order of $0.5 \div 1 \mu m$ at temperatures grater than the bulk Curie temperature by about 10^2 K.

Since discovery of the colossal magneto-resistance much attention has been payed to charge transport in magnetic materials keeping in mind their great potentiality for practical applications. In this regard manganite perovskites are of a particular interest because their magnetic and transport properties are strongly correlated as has been observed [1,2] and explained on the base of the double-exchange model proposed by Zener and de Gennese [3,4]. While in single crystals and high quality epitaxial films of such materials magneto-resistance effects are large in strong magnetic fields of the order of $1T$ close to the Curie temperature, a large low field magnetoresistance has been established to arise in thin films containing interfaces and grain boundaries [5–11,13]. This low field effect appears due to electron spin polarized tunneling [14–19] or spin dependent scattering at grain boundaries/domain walls [11].

Presence of grain boundaries in the sample change not only its magneto-resistance but intrinsic magnetic properties itself as it was observed in Ref. [12,13]. In Ref. [13] grain boundaries were intentionally inserted in a film of $La_{0.7}Sr_{0.3}MnO_3$ that resulted in an increase of the ferromagnet transition temperature by more than $50\ K$ (an increase of the Curie temperature due to a strain caused by the grain boundaries was also reported in [20]).

In this paper we show that the strain field of a grain boundary can result in a significant change of the magnetic structure of a ferromagnet giving rise to a magnetic ordering phase in a large enough region along the dislocation wall at temperatures noticeably higher than the bulk Curie temperature [21].

Grain boundaries in a crystal produce strain fields and elastic deformations that can increase the Curie temperature of a ferromagnet due to the dependence of the exchange energy on the distance between the neighboring atoms. This effect can be particularly large in $La_{1-x}Sr_xMnO_3$ materials where the ferromagnetic ordering is due to the double-exchange ferromagnetic coupling that is extremely sensitive to lattice distortion (see [24] and references there). Hydrostatic pressure p applied to $La_{1-x}Sr_xMnO_3$ $(0.15 \leq x \leq 0.5)$ increases the Curie temperature with a very high pressure coefficient [25,26]:

$$\gamma = \partial \ln T_c / \partial p \approx 0.065 GPa^{-1} \tag{1}$$

In this paper we assume that the grain boundary stress affects ferromagnet properties due to local change of the volume of the crystal as ferromagnet parameters locally depend on the relative volume change (the elastic dilatation) $\epsilon_{ii}(x,y)$; (ϵ_{ik} - the strain tensor). In general, the influence of local changes of ferromagnet parameters on the magnetization is not local due to long-range correlations in the ferromagnet being determined by the minimum condition of the ferromagnet free energy. We find the Curie temperature, the temperature dependence and space distribution of the equilibrium magnetic moment $\mathbf{M}(x,y)$ of a ferromagnet with a tilt grain boundary solving the Landau-Lifshitz equation which is written as follows.

$$-\alpha \frac{\partial^2 M}{\partial \mathbf{x}^2} + 2a\left(T - T_{c_0} - \delta T_c(x,y)\right) M + 4BM^3 = 0 \tag{2}$$

Here α - the exchange constant, a and B are parameters in Ginsburg-Landau expansion of the free energy of a ferromagnet (see [28]) $\mathbf{x} = (x,y,0)$ where y-axis is perpendicular to the grain boundary, T_{c0} is the Curie temperature in the absence of the grain boundary; the local change of the critical temperature in Eq.(2) is

$$\delta T_c = -g T_{c0} \epsilon_{ii}(x,y), \quad g = -\left.\frac{\partial \ln T_{c0}}{\partial \epsilon_{ii}}\right|_{\epsilon_{ii}=0} \approx K\gamma \tag{3}$$

where K is the compression modulus. The boundary conditions for Eq.(2) is finiteness of $M(x, y)$ at the infinity [27].

The dilatation $\epsilon_{ii}(x, y)$, and hence the local temperature shift $\delta T_c(x, y)$ (see Eq.(2,3)) depends on the concrete structure of the boundary.

Boundaries of long-range stress fields. Despite the equilibrium grain boundaries produce short-range stress fields around them, experiments and theory show that boundaries of non-equilibrium configuration can do produce long-range strain fields in the crystal [29]. Below we present elastic dilatation for grain boundaries that may be relevant to the experimental situation of [13,20]

a) For a discontinuous tilt boundaries schematically shown in Fig.(1) the dilatation ϵ_{ii} is as follows [29].

$$\epsilon_{ii}(x, y) = \frac{\epsilon_0}{2} \ln \frac{(x - L/2)^2 + y^2}{(x + L/2)^2 + y^2} \tag{4}$$

where $\epsilon_0 = (b/D)(1 - 2\sigma)/(1 - \sigma)$, D is the distance between neighboring dislocations in the array, σ is the Poisson ratio; $L \gg D$ is the length of the boundary. From Eq.(4) it follows that the elastic dilatation disappears at distances $\sim L \gg D$ (see also Fig.(1)).

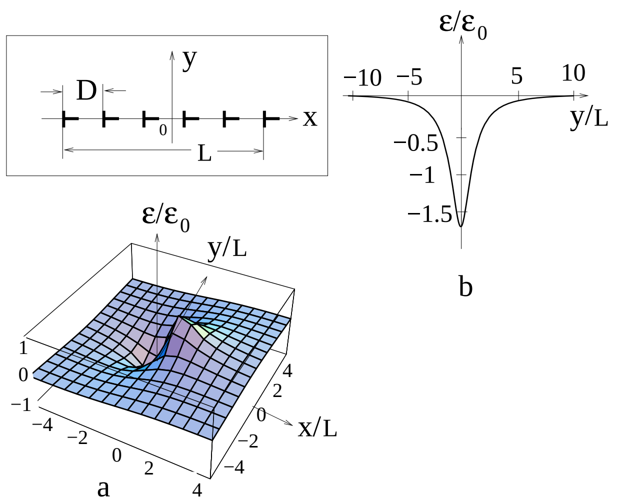

FIG. 1. a) Long-range relative dilatation produced by discontinuous tilt boundary of the length L (shown in the insertion); b) dependence of the relative dilatation on the distance from the boundary at $x = 1.2L$.

b) *A 45° tilt boundary.* In experiment [13,20] artificial boundaries in a cubic crystal were of such a structure that on one side of the boundary ($y < 0$) the [100] crystal axis was parallel to the boundary while on the other side ($y > 0$) the [100] axis was rotated by 45° with respect to the boundary. For the rotated part of the structure ($y > 0$) distortions in such a boundary have two energetically equal directions due to the symmetry of the crystal with respect to the axis perpendicular the boundary plain. In the equilibrium configuration of the boundary the distortion in the boundary corresponds to only one of these two directions and long-range stress fields are absent. However, under the film growth directions of the distortion in different grains along the boundary (nucleated at different points) may have different directions corresponding to the two above-mentioned options that results in a long-range stress field produced by such a boundary the width of which is of the order of the characteristic grain size. We find it using a misfit dislocation model of the boundary as is shown in Fig.(2). Calculations show that in this case the dilatation ϵ_{ii} at distances much greater than the dislocation spacing D in the boundary ($\sqrt{x^2 + y^2} \gg D$) is as follows.

$$\epsilon_{ii}(x, y) = \epsilon_0 \sum_{n=-\infty}^{\infty} (-1)^n \ln \frac{(x - L_n - l_n)^2 + y^2}{(x - L_n)^2 + y^2} \tag{5}$$

where $L_n = \sum_{k=0}^{n} l_n$ and l_n is the length of the boundary inside the n-th grain; these lengths are randomly distributed according to the random distribution of the sizes of the grains. In order to show its main feature we also present here dilatation $\epsilon_{ii}(x, y)$ for a periodic structure of the boundary, assuming the period of it $l_0 \gg D$ to be the characteristic size of the grains (that is $l_n = l_0$); in this case the dilatation is as follows.

$$\epsilon_{ii}(x, y) = \epsilon_0 \ln \frac{1 + 2e^{(-\pi|x|/l_0)} \cos(\pi|y|) + e^{(-2\pi|x|/l_0)}}{1 - 2e^{(-\pi|x|/l_0)} \cos(\pi|y| + e^{(-2\pi|x|/l_0)}} \tag{6}$$

From Eq. (5) and Eq.(6) one sees that such a boundary produces a long-range dilatation that spreads to distances $\sim l^{(0)}$ ($l^{(0)}$ - the characteristic size of the grains along the boundary, see Fig.(2)).

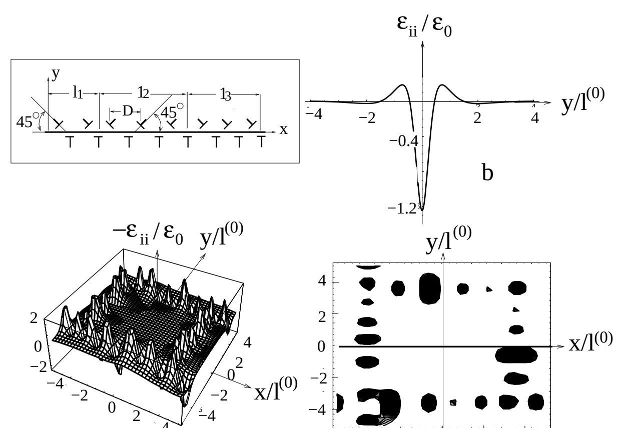

FIG. 2. a) Long-range relative dilatation produced by a non-equilibrium 45° tilt boundary (shown in the insertion); b) dependence of the relative dilatation on the distance from the boundary at $x = -0.75l_0$; c) magnetization regions along the boundary at temperature $(T - T_{c0})/T_{c0} = 0.1$

According to Eq.(3) the dilatation Eq.(15,4,5,6) determine the local change of the Curie temperature $\delta T_c(x, y)$

in Eq.(2). Inserting dimensional variables one sees the solutions of Eq.(2) to be governed by the following dimensionalless parameter

$$\lambda = \left(\frac{d_0}{\xi_0}\right)^2 \frac{\delta T_c^{(0)}}{T_{c0}} \sim \left(\frac{d_0}{\xi_0}\right)^2 K\gamma \tag{7}$$

where $\delta T_c^{(0)}$ and d_0 are the characteristic value and the characteristic variation in space of $\delta T_c(x,y)$, respectively; $\xi_0 = \sqrt{\alpha/(2aT_{c0})}$ is the characteristic correlation length in the ferromagnet at $T = 0$. We solve the non-linear equation Eq.(2) for a general situation in two limiting cases $\lambda \ll 1$ and $\lambda \gg 1$ specifying the form of the $\delta T_c(x,y)$ afterwards.

1) For the case $\lambda \ll 1$ we assume $\epsilon_{ii}(x,y)$ to be localized along the boundary (decaying in the y-direction) and to be a periodic function in the x-direction with the period d_0. Under this assumption we solve Eq.(2) in the following way.

Expanding the local critical temperature change and the magnetization in the Fourier series

$$\begin{array}{l} \delta T_c(x,y)/\delta T_c^{(0)} = \sum_{n=-\infty}^{\infty} V_n(y)\exp(i2\pi nx/d_0), \\ m(x,y) = M(x,y)/M_0 = \\ \sum_{n=-\infty}^{\infty} A_n(y)\exp(i2\pi nx/d_0) \end{array} \tag{8}$$

($M_0 = \sqrt{a/2B}$ is the magnetization at $T = 0$) and inserting it in Eq.(2) one sees that in the first non-vanishing approximation in λ, the differential equation for any A_n with $n \neq 0$ is a linear differential equations of the second order with constant coefficients and the right side equal to $\lambda V_n(y)A_0(y)$ (that is they are small comparing with A_0: $A_n(y) \sim \lambda A_0$). Solving these equations with the above-mentioned boundary conditions one gets the following non-linear equations for the zero harmonic A_0 of the magnetization m.

$$\frac{\partial^2 A_0}{\partial \zeta^2} - \lambda\left(V_{eff}(\zeta) + E\right)A_0 - CA_0^3 = 0; \tag{9}$$

where $\zeta = y/d_0$, constant $C = (d_0/\xi_0)^2$, "energy" $E = (T - T_{c0})/\delta T_c^{(0)}$; the effective "potential" $V_{eff}(\xi)$ is as follows.

$$V_{eff}(\zeta) = -\lambda \sum_{n=1}^{\infty} \frac{1}{n} V_n(\zeta) \int_{\infty}^{\infty} V_n^*(\zeta + \zeta')e^{-n|\zeta'|}d\zeta' \tag{10}$$

While writing Eq.(9) we used the relations $V_0 = 0$, $V_n = -V_{-n} = -V_n^*$ which are valid for the case of our interest $\delta T_c(x,y) = -\delta T_c(x,-y)$. According to Eq.(9), for $\lambda \ll 1$ function $A_0(\zeta)$ varies at a distance that is much grater than the interval where $V_{eff}(\zeta)$ is localized. Therefore, one may solve Eq.(9) in the region $\zeta \gg 1$ (where V_{eff} can be neglected) with the boundary condition

$$\left.\frac{dA_0}{d\xi}\right|_{\xi=1} - \left.\frac{dA_0}{d\xi}\right|_{\xi=-1} = \lambda \int_{-\infty}^{\infty} V_{eff}(\xi)d\xi A_0(0) \tag{11}$$

Non-linear Eq.(9) with boundary condition Eq.(11) has two bifurcation points: 1) at at the bulk critical temperature $T = T_{c0}$, ($E = 0$), and at a new critical temperature $T_c = T_{c0} + \Delta T_c > T_{c0}$ ($E = E_c = \Delta T_c/\delta T_c^{(0)}$) at which a spontaneous magnetization arises around the grain boundary, where $\Delta T_c = (\lambda \delta T_c^{(0)}/4) < V_{eff} >^2$. Solving Eq.(9) and Eq.(11) one gets the magnetization to be as follows.

$$m(y) = \sqrt{2(T_c - T)(T - T_{c0})/T_{c0})}/ \left(\sqrt{T - T_{c0}}\cosh(y/\xi_0(T)) + \sqrt{Tc - T_{c0}}\sinh(|y|/\xi_0(T)\right) \tag{12}$$

for $T_{c0} \leq T \leq T_c$, and

$$m(y) = \left(\frac{T_{c0} - T}{T_{c0}}\right)^{1/2} \frac{1 + a\exp\left(-\sqrt{2}|y|/\xi_0(T)\right)}{1 - a\exp\left(-\sqrt{2}|y|/\xi_0(T)\right)} \tag{13}$$

for $T < T_{c0}$, where

$$a = \sqrt{1 + 2\frac{T_{c0} - T}{T_c - T_{c0}}} - \sqrt{2\frac{T_{c0} - T}{T_c - T_{c0}}} \tag{14}$$

and the temperature dependent correlation length is $\xi_0(T) = \xi_0\sqrt{T_{c0}/|T - T_{c0}|}$

Therefore, for $\lambda \ll 1$ the grain boundary enhanced magnetization in such a way that in the temperature region $T_{c0} \leq T \leq T_C$ there is a sheet of magnetic ordering along the boundary the width of which is $\sim \xi_0(T)$ as shown in Fig.3

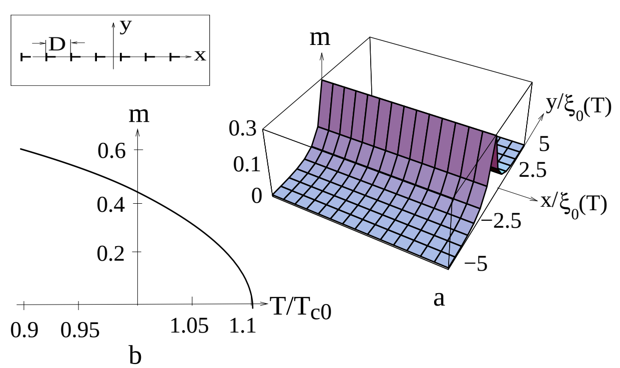

FIG. 3. a) Relative magnetization at $(T - T_{c0})/T_{c0} = 1.05$ and $(T_c - T_{c0})/T_{c0} = 1.1$ for $\lambda \ll 1$; b) temperature dependence of the magnetization at the boundary ($y = 0$)
. The insertion shows a low-angle tilt boundary.

For example, the dilatation $\epsilon_{ii}(x,y)$ produced by a low-angle tilt boundary (see the insertion in Fig.(3)) is [29])

$$\epsilon_{ii}(x,y) = -\epsilon_0 \frac{\sin(2\pi x/D)}{\cosh(2\pi y/D) - \cos(2\pi x/D)}, \tag{15}$$

and using Eq.(15, 10) and Eq.(8) one finds $< V_{eff} >= -1.72\lambda$ and the new Curie temperature T_c to be

$$T_c = T_{c0}(1 - 0.43\lambda^3 g\epsilon_0) \approx T_{c0}(1 + 0.43\gamma\lambda^3 K\epsilon_0) \quad (16)$$

For the case $\lambda \gg 1$ the pattern of the magnetization temperature dependence differs and may be analyzed as follows.

The Curie temperature T_c is found as the lowest level E_0 of the "energy" $E = T - T_{c0}$ of the linearized Eq.(2) [22]. For $\lambda \gg 1$ it can be easily found because the eigenfunction corresponding to this lowest level should have no nodal lines [30] while wave functions of the linearized Eq.(2) oscillate at distances $\delta r \sim \lambda^{-1} d_0$. It means that the lowest level should be so close to the bottom of the "potential well" that the ground state function is localized around the point of the minimum of it $\mathbf{x}_0 = (x_0, y_0)$ at distances $|\mathbf{x} - \mathbf{x}_0| \sim \lambda^{-1} d_0 \ll d_0$ (where only one oscillation of the ground state wave function takes place), and hence the Taylor series expansion of $\delta T_c(x, y)$ in Eq.(2) is possible. In this case calculations show that magnetic ordering appears in a narrow tube of the width $\sim \lambda^{-(1/4)} d_0$ around $\mathbf{x}_0$ at the temperature $T = T_c + \delta T_c$ where the shift of the critical temperature is $\delta T_c \approx T_{c0}|g|\epsilon_0 \sim K\gamma\epsilon_0 T_{c0}$.

With a further decrease of temperature the region of the magnetization expands and for $T_c < T < T_{c0}$, according to Eq.(2), the magnetization of the ferromagnet in the presence of the boundary can be written as

$$m(x, y) = \sqrt{1 - g\epsilon_{ii}(x, y) - T/T_{c0}} \quad (17)$$

As follows from Eq.(17) and Eq.(4, 5, 6), in the regions where $\epsilon_{ii} < 0$, spontaneous magnetization arises along the boundary at distances of the order of the grain size at temperatures exceeding the bulk temperature T_{c0} (see Fig.(2)). With further decrease of the temperature, in the temperature interval where $\xi_0(T) \gg l_0$, the magnetization is described by Eq.(12, 13) (see also Fig.(3) with $d_0 = l_0$.

Using Eq. (3) and Eq. (1,5) one estimates the increase of the Curie temperature due to the long-range boundary strain to be $\delta T_c^{(0)}/T_{c0} \approx \gamma K(b/D)$ (K is the compressibility). For the experimental value of $\gamma = 0.065 GPa$ [25,26], T_{c0} =350 K, $D \approx 10b$, and typical values $K = 50 GPa$ one gets $\delta T_c^{(0)} \approx 100$K while the characteristic size of the regions of the spontaneous magnetization at $T_{c0} < T < T_{c0} + \delta T_c^{(0)}$ being $l^{(0)} \sim 0.1 \div 1 \mu m$ for the grain size typical for the experiment.

A.K acknowledges the hospitality of the Department of Applied Physics and the Department of Microelectronics and Nanoscience, Chalmers University of Technology and Göteborg University.

The study was supported by SSF OXIDE and SSF QUANTUM DEVICES and NANOSCIENCE programs.

*E-mail:
kadig@fy.chalmers.se and kadig@kam.kharkov.ua

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